

**Argonne National Laboratory**

**A CRITICAL COMPARISON OF MEASURED  
AND CALCULATED FISSION RATIOS  
FOR ZPR-III ASSEMBLIES**

**by**

**W. G. Davey**

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ARGONNE NATIONAL LABORATORY  
9700 South Cass Avenue  
Argonne, Illinois

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by

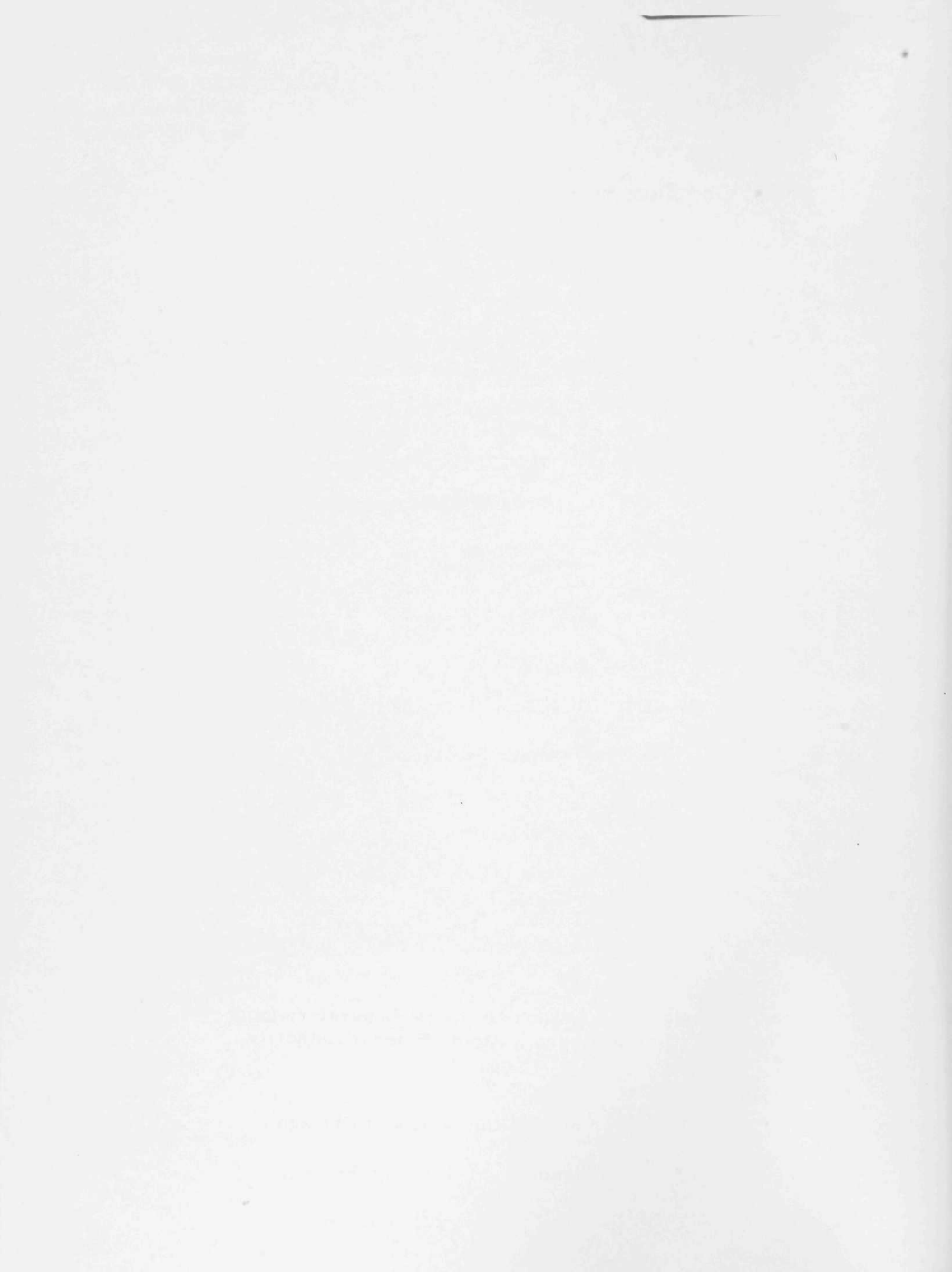
W. G. Davey\*

Idaho Division

September 1962

\* Assigned to Argonne National Laboratory from  
the United Kingdom Atomic Energy Authority

Operated by The University of Chicago  
under  
Contract W-31-109-eng-38  
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# A CRITICAL COMPARISON OF MEASURED AND CALCULATED FISSION RATIOS FOR ZPR-III ASSEMBLIES

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## ABSTRACT

A critical comparison of measured and calculated central fission ratios for 18 ZPR-III fast reactor assemblies has been made with the object of examining the accuracy of computation of spectra and of the fission cross sections used. This comparison uses fission ratios measured with Kirn absolute fission chambers and computed with ANL cross-section Set 635.

The Kirn chambers and experimental technique are briefly described. It is shown that ratios measured with threshold detectors must be corrected for the effects of inelastic scattering in the chamber walls. Possible sources of error in the experimental technique are discussed and experimental evidence for the validity of the method is presented.

The derivation of ANL Set 635 is briefly described. It is shown that Set 635 is a modification of the Yiftah, Okrent, and Moldauer ANL Set 135, and that the central spectra and fission ratios calculated with the two sets are generally similar. The  $U^{234}$  and  $U^{236}$  fission cross sections are not given in Sets 135 and 635; these have been taken from ANL Set 179.

The measured and calculated fission ratios obtained with  $U^{233}$ ,  $U^{234}$ ,  $U^{235}$ ,  $U^{236}$ ,  $U^{238}$ ,  $Pu^{239}$ , and  $Pu^{240}$  are compared, both to search for trends which might occur with progressive changes in spectra, and also to determine the accuracy of prediction of ratios.

It is found that the calculated relative fission rates of  $Pu^{239}$  and  $U^{233}$  are within about  $\pm 1\frac{1}{2}\%$  of the measured values, and the calculated rates for  $Pu^{240}$ ,  $U^{236}$ , and  $U^{238}$  relative to either  $Pu^{239}$  or  $U^{233}$  are within  $\pm 3\frac{1}{2}\%$  to  $\pm 5\%$  of the measured values. However, calculated fission rates for  $U^{235}$  and  $U^{234}$  are about 6% low and 8% high, respectively, relative to those of the other five isotopes.

If the experimental data are correct, then it appears that the assumed fission cross sections of  $Pu^{239}$ ,  $Pu^{240}$ ,  $U^{233}$ ,  $U^{236}$ , and  $U^{238}$  are fairly accurate, but that the assumed cross sections of  $U^{235}$  and  $U^{236}$  are, respectively, 6% low and 8% high. It is emphasized that, even if only some of the chambers give erroneous results, radically different conclusions could be reached and the importance of checking on the intercalibration of the fission chambers is argued.

## 1. INTRODUCTION

The comparison of measured and calculated reaction rates in reactors has long been a recognized method of examining the accuracy of nuclear data and computation techniques. This analysis has frequently been applied to fast reactors, two of the most recent studies being those of Yiftah, Okrent, and Moldauer (YOM)<sup>(1)</sup> and of Long *et al.*<sup>(2)</sup> In both of these studies the fission ratios measured in the extensive series of ZPR-III fast reactor assemblies were examined, but in neither case was there a detailed evaluation of the data. In addition, the data used were partially in error, as Davey and Curran<sup>(3)</sup> have shown (confirmed in recent ZPR-III measurements) that the measured reaction rates of threshold fissile materials are significantly affected by inelastic scattering in the walls of the fission chambers.

Since the effects of inelastic scattering significantly altered the measured data and since there were also other minor inaccuracies in the measured ratios, the present study was undertaken.

In order to minimize the spread of experimental data, only the fission ratios measured with the Kirn absolute fission chambers<sup>(4)</sup> were used. The early, less accurate measurements with fission chambers calibrated by thermal irradiation<sup>(5)</sup> and the radiochemical ratios<sup>(5)</sup> have not been included.

The calculated spectra and fission ratios (with some slight changes) were obtained with cross-section set ANL Set 635, which has recently been derived by the author from the Yiftah, Okrent, Moldauer ANL Set 135 and used in a study of the critical sizes of 22 ZPR-III assemblies.<sup>(6)</sup> It should be noted that Davey's data were used because they covered a large range of ZPR-III assemblies and not because the spectra were expected to be any more accurate than those derived with other cross-section sets. In particular, the central spectra calculated with Set 635 and Set 135 are closely similar although critical sizes calculated with the two sets can differ appreciably.

## 2. MEASUREMENTS WITH KIRN ABSOLUTE FISSION CHAMBERS

### 2.1 Description of the Chambers

The chambers have been fully described by Kirn and will only be described briefly here. A section through a Kirn chamber is given in Fig. 1 together with those of two similar, gas-flow chambers.

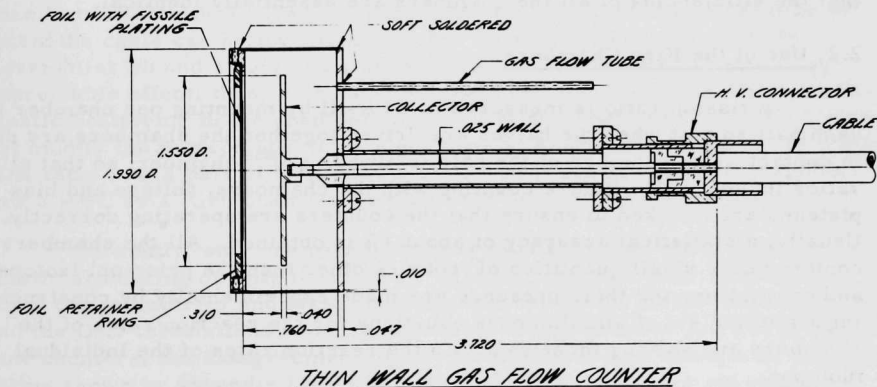
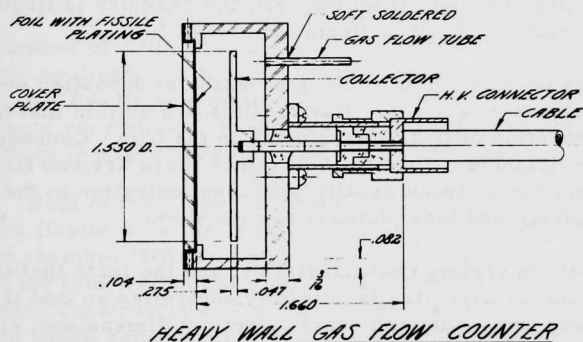
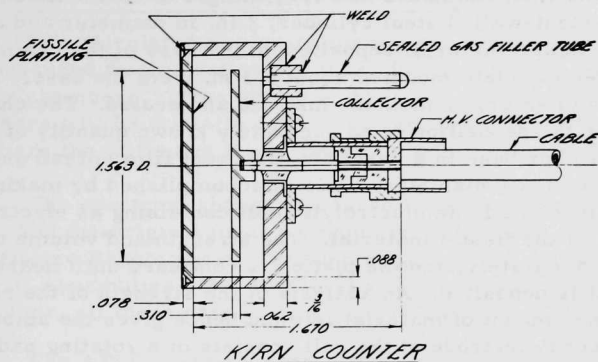


Fig. 1. Absolute Fission Chambers

Basically, the Kirn chambers are very simple in construction. The chamber body is a stout-walled steel cylinder, 2 in. in diameter and about 1 in. high, with the fissile material deposited on the base of the chamber and a circular collection plate mounted about 0.3 in. from the base. The chamber is filled with an argon-methane mixture and sealed. The chamber is made absolute by the deposition of an accurately known quantity of fissile material on the chamber base in a very thin, adherent film spread over a circle of a precisely known diameter. This is accomplished by making the chamber base an electrode in an electrolytic cell containing as electrolyte a solution of a salt of the fissile material. The strength and volume of the solution are known accurately, and the plating is continued until nearly all the fissile material is deposited. An analysis of the strength of the residual electrolyte gives the amount of material left and hence gives the amount deposited. The second electrode of the cell consists of a rotating paddle which stirs the electrolyte, and consequently the fissile material is deposited as a uniform film. After removal from the cell, the chamber is fired to reduce the fissile material to a hard, uniform oxide film.

The chambers contain 100 to 1000  $\mu\text{g}$  of material deposited over an area about 1 in. in diameter, and these fissile films are so thin that there is essentially no absorption of fission fragments in the film. Consequently, the chambers have efficiencies close to 100% (since there are two fission fragments per fission, one of these usually produces ionization in the chamber), and the voltage and bias plateaus are excellent.

These excellent operating characteristics, and the facts that the area over which the material is deposited is carefully controlled so that it is constant and the chambers are made with nearly identical dimensions, ensure that the efficiencies of all the chambers are essentially identical.

## 2.2 Use of the Kirn Chambers

A fission ratio is measured in ZPR-III by mounting one chamber in each half so that when the halves are driven together the chambers are nearly in contact. One chamber of the pair is always a  $\text{U}^{235}$  chamber, so that all ratios include  $\text{U}^{235}$ . Before counting with the chambers, voltage and bias plateaus are checked to ensure that the counters are operating correctly. Usually, a statistical accuracy of about 1% is obtained. All the chambers contain fairly small quantities of isotopes other than the principal isotope, and corrections for their presence are made experimentally by constructing a suitable set of simultaneous equations for the reaction rates of the chambers and solving these to obtain the reaction rates of the individual isotopes.

The ZPR-III assemblies are constructed of plates, usually  $\frac{1}{8}$  in. thick, and consequently are not homogeneous. In order that the measured fission ratios should be characteristic of a homogeneous system, the fission chambers are mounted with the fissile material perpendicular to the plates so that heterogeneities of spectrum tend to average out.

### 2.3 Inelastic Scattering in the Fission Chamber Walls

The calculated fission ratios are for homogeneous reactors, whereas the measured fission ratios are those for the neutron spectrum inside the fission chambers. If the chamber walls are thin, the spectrum inside the chambers will be trivially different from that outside, but with the Kirn chambers the walls are thick enough to modify the spectrum significantly.

The spectrum change is caused by inelastic scattering in the chamber walls, and the original measurements by Davey and Curran using the Argonne Fast Source Reactor have now been supplemented by measurements in ZPR-III Assemblies 35(7) and 38(8) and by a number of DSN calculations by the author, all these data being given in Appendix I.

The AFSR measurements were made by placing cylindrical shells of steel around long, thin fission chambers, and the DSN calculations investigated the effect of thin, spherical iron shells on the spectrum at the center of a number of ZPR-III assemblies, so that in neither case are the results directly applicable to the Kirn fission chambers. However, the measurements in ZPR-III Assemblies 35 and 38 were made with gas-flow fission chambers very similar to the Kirn chambers. The two types of chambers are shown in Fig. 1 together with a Kirn chamber.

Both of these special chambers are of the gas-flow type in order that the fissile foils they contain can be easily demountable. The heavy-walled chamber (HWC) is otherwise closely similar to the Kirn chamber, whilst the thin-walled chamber (TWC) contains the minimum amount of steel and other constructional material. The TWC also possesses a metallic extension which removes the hydrogen-containing cable 2 in. further from the chambers than with the HWC or Kirn chambers, so that the moderating effect of the cable can be investigated. The measurements with ZPR-III Assemblies 35 and 38 give no significant evidence that the cable has any appreciable effect; thus, it is assumed that any effects are due solely to inelastic scattering in the chamber walls. It is considered that the TWC will not modify the neutron spectrum significantly; hence, measurement of a fission ratio with a pair of foils in two TWC's and then in two HWC's shows how a HWC or a Kirn chamber modifies the neutron spectrum.

Currently, only one pair of foils is available, and the effect of inelastic scattering on only the  $U^{238}/U^{235}$  fission ratio has been investigated. Since the geometries and steel shell thicknesses in the AFSR experiments and the DSN calculations were considerably different from those of the gas-flow counters, the disagreement of the actual percentage changes in fission ratios shown in Appendix I is to be expected. However, it is reasonable to expect that, as we are considering fairly small changes, all effects should be fairly linear, and the AFSR and DSN data give the relative changes fairly accurately. Hence, the measurements on the  $U^{238}/U^{235}$  ratio with the gas-flow chambers can normalize the AFSR measurements and the DSN calculations so that corrections can be made for all the fission ratios measured with the Kirn chambers.

The percentage changes in the  $U^{238}/U^{235}$  ratio which were measured in Assemblies 35 and 38 were both close to 8%, and the AFSR and DSN data were normalized to this value. These data are presented in Table I.

Table I  
ESTIMATES OF INELASTIC SCATTERING CORRECTIONS TO  
KIRN FISSION CHAMBER MEASUREMENTS

Percentage Change <sup>(a)</sup>								
Fission Ratio	AFSR Measurements	DSN Calculations				ZPR-III Measurements		Final Assumed Values
		Assy. 14	Assy. 25	Assy. 32	Assy. 32	Assy. 35	Assy. 38	
		(S4)	(S4)	(S4)	(S8)			
$\frac{U^{233}}{U^{235}}$	(b)	-0.10	0.03	-0.07	-0.07	(b)	(b)	Zero
$\frac{Pu^{239}}{U^{235}}$	$0 \pm \frac{1}{2}$	0.5	0.6	0.5	0.5	(b)	(b)	Zero
$\frac{Pu^{240}}{U^{235}}$	(b)	3.4	3.8	3.4	3.4	(b)	(b)	$4 \pm 1$
$\frac{U^{234}}{U^{235}}$	$5 \pm 1$	3.2	3.5	3.2	3.2	(b)	(b)	$4 \pm 1$
$\frac{U^{236}}{U^{235}}$	(b)	5.9	6.1	5.8	5.8	(b)	(b)	$6 \pm 1\frac{1}{2}$
$\frac{U^{238}}{U^{235}}$	8.0 <sup>(c)</sup>	8.0 <sup>(c)</sup>	8.0 <sup>(c)</sup>	8.0 <sup>(c)</sup>	8.0 <sup>(c)</sup>	$8.3 \pm 1.0$	$7.4 \pm 1.5$	$8 \pm 2$

(a) A positive sign indicates that the fission ratio measured with Kirn chambers must be increased to give the true value.

(b) Not measured or not calculated.

(c) Normalized to this value.

It can be seen that the DSN calculations give mutually consistent results, indicating that the corrections are fairly independent of the type of assembly. The DSN results are also in reasonable agreement with the AFSR data.

The final column of Table I gives the values of the corrections to be applied to all the measurements with Kirn chambers. Fairly large errors have been assumed to allow for the numerous uncertainties in their estimation.

#### 2.4 Validity of the Fission Chamber Measurements

Before comparing measured and calculated fission ratios, we will consider the possible sources of error involved in the measurements and

examine their validity. In this context we will not discuss the question of inelastic scattering in the chamber walls already covered in Section 2.3.

Considering the manufacture and use of the chambers we can list the following sources of error:

In manufacture we have

- (a) uncertainty in the quantity of fissile material deposited in the chamber;
- (b) uncertainty in its isotopic composition; and
- (c) the possibility of different counting efficiencies in the chambers due to different internal geometries or non-uniformity of the fissile film.

In use we have the possibilities of

- (d) deterioration of the fissile film;
- (e) deterioration of the gaseous filling due to leakage of air; and
- (f) operational errors, such as poor selection of operating bias.

With regard to the comparison with calculations, we also have the question

- (g) Do the chambers measure the average spectrum in the reactor?

These questions were, of course, carefully examined by Kirn, but they have been extensively considered also by many others, particularly by C. C. Miles, ANL, Idaho Division, who has manufactured some Kirn-type chambers.

With regard to (a) and (b), there have been a number of independent analyses of the strength of the initial electrolyte, of the strength of the residual electrolyte, and isotopic analyses plus  $\alpha$ -counting of the deposited materials, so that there can be no doubt that the techniques involved are reliable and accurate. This does not, of course, remove the possibility of, for example, an inadvertent error in the volume of electrolyte used.

There is some experimental evidence regarding (c) which also throws some light upon (a) and (b). The Kirn chambers were made identically as far as possible, and thus it is very probable that they have the same efficiency.

However, on a number of occasions a pair of similar Kirn chambers have been irradiated simultaneously in ZPR-III and their counting rates compared with those expected from the quantity of fissile material present. In addition,  $U^{238}/U^{235}$  fission ratios have been measured in some assemblies both with Kirn counters and with gas-flow counters manufactured some years later by C. C. Miles with electrolytes of different isotopic composition and strength. These data (which are detailed in Appendix II) show:

1. that the relative count rates of the Kirn chambers which were tested are in excellent agreement with those expected from their weights;
2. that the  $U^{238}/U^{235}$  fission ratios measured with the Kirn and Miles chambers are in excellent agreement, thus giving great confidence that these measurements are correct.

We therefore conclude that the Kirn chambers can give valid measurements of relative fission rates.

With regard to (d), examination of the relative count rates of similar chambers over a considerable period of time (for example, the  $U^{235}$  Kirn chambers numbers 4 and 5) show no evidence of the deterioration of the fissile foils.

Regarding (e) and (f), examination of measured ratios shows that it is probable that these errors do occur occasionally without being detected by the experimenters. These occurrences are rare, however, since the excellent operating characteristics of the chambers make detection of a defective counter a relatively simple matter. When the deterioration is slight, a poor measurement may be accepted, but then the fission ratio is only slightly in error and even then the error can possibly be detected by a systematic examination of a range of fission ratios. This procedure is, in fact, followed in Section 4 of the present work, in which certain deviant data are rejected as being highly improbable statistically.

The last uncertainty, (g), is possibly the most difficult to examine. A limited amount of experimental information on the effects of local changes in environment on the measured  $U^{238}/U^{235}$  fission ratio has been obtained in ZPR-III Assembly 38.<sup>(8)</sup> These apparently show that some considerable local changes do not affect the measured ratio, and this implies that it is characteristic of the reactor as a whole. However, there are some discordant data, and this information is inadequate to establish any general conclusions. Measurements with fissile foils<sup>(5)</sup> and calculations<sup>(9)</sup> show that there can be considerable local variations in fission rates, but the Kirn chambers presumably measure some sort of average spectrum. The degree of uncertainty involved cannot be established at present, but it is probably unlikely to exceed a few percent. This must still be regarded as dubious until more information is available.



The general conclusion of this section is therefore that the Kirn chambers do give valid measurements of fission ratios (if the inelastic scattering correction is included), but some deviant results are to be expected because of slight chamber deterioration. The heterogeneous nature of ZPR-III assemblies introduces an additional uncertainty, probably small.

## 2.5 Experimental Values of Fission Ratios

The present work has considered Kirn measurements in 19 ZPR-III assemblies, of which numbers 24 and 38 have identical cores which should give essentially identical ratios. Therefore, 18 cases are examined.

The data for Assemblies 10, 11, 12, 14, 16, and 17 have generally been taken from reference (5), for Assemblies 20, 23, 24, 25, 32, and 33 from reference (2), and for 29, 30, 31, 34, 35, 36, and 38 from references (10), (11), (12), (13), (7), (14), and (8).

The  $U^{233}/U^{235}$  ratios are incorrectly quoted in (5), and those given (correctly) by Yiftah, Okrent, and Moldauer<sup>(1)</sup> have been used. In addition, some quoted data for  $U^{236}/U^{235}$  ratios are based on a preliminary isotopic analysis and are slightly erroneous. These data have been corrected by the author.

All experimental data are given in Section 4 of this report.

## 3. CALCULATION OF CENTRAL FISSION RATIOS

The central spectra were calculated by use of the 16-group ANL cross-section Set 635 together with the DSN neutron transport code in the S4 approximation. The calculations were made in a recent study by the author<sup>(6)</sup> of the critical sizes of ZPR-III assemblies and were made on spherical versions of these assemblies.

ANL Set 635 consists of the 16-group Yiftah, Okrent, and Moldauer ANL Set 135 with modified values of  $\alpha$  and  $\nu$  for  $U^{235}$  and of  $\sigma$  (transport) and  $\sigma$  (elastic removal) for aluminum, iron, nickel, and chromium. The differences between Sets 135 and 635 are not such as to cause large differences in central spectra, and a comparison between the present fission ratios and those given<sup>(2)</sup> by Set 135 shows that they generally differ by not more than 2% to 4%. The  $U^{236}/U^{235}$  ratios differ by considerably more, but this is probably largely due to a different choice of nuclear data for  $U^{236}$ . Neither Set 135 nor Set 635 includes nuclear data for  $U^{234}$  and  $U^{236}$ , and in this study the fission cross sections for these nuclides have been taken from ANL Set 179, which has the same group structure as Sets 135 and 635. It should be noted that the author used different data for  $U^{234}$  and  $U^{236}$  in the study of critical size, but these were rather crudely estimated and the Set 179 values are believed to be superior. These are given in Appendix III.

The central spectra, average cross sections, and fission ratios are detailed in Appendix IV. The fission ratios are also tabulated in Section 4.

#### 4. COMPARISON OF MEASURED AND CALCULATED RATIOS

##### 4.1 Method

Each set of fission ratios was examined in the following way:

- (a) The calculated and measured ratios (not corrected for inelastic scattering) were tabulated in the order of increasing calculated ratio, and the ratio of calculated to measured fission ratio ( $C/E$ ) was obtained.
- (b) The first half and second half of each list were then analyzed separately. The average  $C/E$  for each half was obtained and compared to see if there were any trend in  $C/E$  with progressive change in spectrum since, if present, this would indicate a systematic error in assumed fission cross sections or calculated spectra. This analysis included rejection of erroneous values of  $C/E$ , the criterion for rejection being a deviation of more than three standard deviations from the mean of the remainder of the set. If there was no significant difference between the first half and second half of a set, then the average of the complete set and the accuracy of its determination was obtained.
- (c) The mean value of  $C/E$  was then corrected for inelastic scattering in the chamber walls by means of the data of Table I. These corrected values of  $C/E$  were then examined for trends.

##### 4.2 Detailed Comparison of Ratios

The details of the comparison of the experimental fission ratios (not corrected for inelastic scattering in the chamber walls) and the calculated values are given in Tables II through VII. It can be seen that in only one case, that of the  $Pu^{239}/U^{235}$  ratio, is there any evidence that the relative values of the calculated and measured ratios are a function of the hardness of spectrum, and even in this case it is dubious that this is statistically significant.

The inelastic scattering correction is applied to the best mean values of the ratio  $C/E$ , i.e., (calculated fission ratio)/(experimental fission ratio), in Table VIII. The corrected values which would be obtained if the fission ratios had been taken relative to  $Pu^{239}$  are presented in Table IX. These values are obtained (except for that for the  $U^{235}/Pu^{239}$  which is the inverse of the  $Pu^{239}/U^{235}$  value of Table VIII) by dividing by the  $Pu^{239}/U^{235}$  value in Table VIII.

Table II

COMPARISON OF EXPERIMENTAL AND CALCULATED  
Pu<sup>239</sup>/U<sup>235</sup> FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experi- mental Ratio (E)	C/E	Analysis of Halves of Set		Analysis of Whole Set	
				Average C/E	Deviation (D)	Average C/E	Deviation (D)
14	1.122	1.05	1.069	1.069	0.000	1.058	+0.011
35	1.145	1.09	1.050		-0.019		-0.008
29	1.147	1.06	1.082		+0.013		+0.024
34	1.153	1.07	1.078		+0.009		+0.020
17	1.156	1.08	1.070		+0.001		+0.012
12	1.181	1.10	1.074		+0.005		+0.016
30	1.201	1.12	1.072		+0.003		+0.014
20	1.217	1.15	1.058		-0.011		0.000
25	1.222	1.17	1.044	1.049	-0.005		-0.014
24	1.226	1.16	1.057		+0.008		-0.001
31	1.231	1.18	1.043		-0.006		-0.015
11	1.242	1.17	1.062		+0.013		+0.004
32	1.244	1.20	1.037		-0.012		-0.021
36	1.250	1.19	1.050		+0.001		-0.008
33	1.251	1.21	1.034		-0.015		-0.024
10	1.262	1.22	1.034		-0.015		-0.024
23	1.273	1.18	1.079		+0.030		+0.021
				Mean C/E		Error on Mean C/E $[\sum D^2/n(n-1)]^{1/2}$	
First 8				1.069		0.004	
Last 9				1.049		0.005	
17				1.058		0.004	
						Error on Individual Values $[\sum(D^2/n)]^{1/2}$	
First 8						0.010	
Last 9						0.014	
17						0.015	

There is some evidence of a trend with spectrum but it is possibly not statistically significant. We therefore assume

$$\text{Best value of } C/E = 1.058 \pm 0.004$$

Table III

COMPARISON OF EXPERIMENTAL AND CALCULATED  
 $U^{233}/U^{235}$  FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experi- mental Ratio (E)	C/E	Analysis of Halves of Set		Analysis of Whole Set	
				Average C/E	Deviation (D)	Average C/E	Deviation (D)
14	1.501	1.45	1.035	1.051	-0.016	1.049	-0.014
17	1.532	1.46	1.049		-0.002		0.000
29	1.543	1.47	1.050		-0.001		+0.001
34	1.545	1.45	1.066		+0.015		+0.017
12	1.548	1.46	1.060		+0.009		+0.011
30	1.559	1.49	1.046		-0.005		-0.003
23	1.559	1.48	1.053		+0.002		+0.004
31	1.569	1.52	1.032	1.046	-0.014		-0.017
20	1.571	1.52	1.034		-0.012		-0.015
33	1.574	1.51	1.042		-0.004		-0.007
32	1.576	1.51	1.044		-0.002		-0.005
36	1.584	1.47	1.078		+0.032		+0.029
24	1.586	1.44	1.101(a)		(a)		(a)
10	1.587	1.52	1.044		-0.002		-0.005
11	1.587	1.51	1.051		+0.005		+0.002
				<div> <div>Mean C/E</div> <div>Error on Mean C/E <math>[\sum D^2/n (n-1)]^{1/2}</math></div> <div>Error on Individual Values <math>[\sum (D^2/n)]^{1/2}</math></div> </div>			
First 7				1.051	0.004	0.009	
Last 7 (excl'd. 24)				1.046	0.006	0.015	
14				1.049	0.004	0.012	

There is no evidence of a trend with spectrum.

$$\text{Best value of } C/E = 1.049 \pm 0.004$$

(a) Rejected as being more than three standard deviations from the mean.

Table IV  
COMPARISON OF EXPERIMENTAL AND CALCULATED  
 $U^{234}/U^{235}$  FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experi- mental Ratio (E)	C/E	Analysis of Halves of Set		Analysis of Whole Set	
				Average C/E	Deviation (D)	Average C/E	Deviation (D)
35	0.283	0.232	1.220	1.186	+0.034	1.179	+0.041
29	0.292	0.259	1.127		-0.059		-0.052
34	0.303	0.247	1.227		+0.041		+0.048
25	0.326	0.253	1.289		+0.103		+0.110
24	0.334	0.246	1.358(a)		(a)		(a)
12	0.342	0.293	1.167		-0.019		-0.012
17	0.342	0.298	1.148		-0.038		-0.031
20	0.349	0.300	1.163		-0.025		-0.016
14	0.350	0.305	1.148		-0.038		-0.031
16	0.350	0.297	1.178	1.173	+0.005		-0.001
30	0.351	0.301	1.166		-0.007		-0.013
11	0.364	0.299	1.217		+0.044		+0.038
31	0.383	0.334	1.147		-0.026		-0.032
36	0.385	0.312	1.234		+0.061		+0.055
32	0.399	0.367	1.087		-0.086		-0.092
10	0.405	0.331	1.224		+0.051		+0.045
33	0.414	0.370	1.119		-0.054		-0.060
23	0.477	0.402	1.187		+0.014		+0.008
				Mean C/E	Error on Mean C/E $[\sum D^2/n(n-1)]^{1/2}$	Error on Individual Values $[\sum (D^2/n)]^{1/2}$	
First 8 (excl'd. 24)				1.186	0.019	0.049	
Last 9				1.173	0.017	0.050	
17				1.179	0.012	0.049	

There is no evidence of a trend with spectrum.

$$\text{Best value of } C/E = 1.179 \pm 0.012$$

(a) Rejected as being more than three standard deviations from the mean.

Table V  
COMPARISON OF EXPERIMENTAL AND CALCULATED  
 $\text{Pu}^{240}/\text{U}^{235}$  FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experimental Ratio (E)	C/E	Analysis of Whole Set	
				Average C/E	Deviation (D)
35	0.288	0.250	1.152	1.102	+0.050
29	0.300	0.289	1.038		-0.064
34	0.311	0.271	1.148		+0.046
20	0.356	0.332	1.072		-0.030
36	0.391	0.337	1.160		+0.058
31	0.394	0.313	1.259(a)		(a)
32	0.410	0.382	1.073		-0.029
33	0.427	0.400	1.068		-0.034

There is insufficient data to look for trends with spectrum.

	Mean C/E	Error on Mean C/E $[\sum D^2/n(n-1)]^{1/2}$	Error on Individual Values $[\sum (D^2/n)]^{1/2}$
Mean of 7 (excl. 31)	1.102	0.020	0.050

Best value of C/E = 1.102 ± 0.020

(a) Rejected as being more than three standard deviations from the mean.

Table VI  
COMPARISON OF EXPERIMENTAL AND CALCULATED  
 $\text{U}^{236}/\text{U}^{235}$  FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experimental Ratio (E)	C/E	Analysis of Whole Set	
				Average C/E	Deviation (D)
29	0.0931	0.082	1.135	1.129	+0.006
34	0.0949	0.076	1.249(a)		(a)
36	0.112	0.094	1.191		+0.062
30	0.112	0.099	1.131		+0.002
31	0.120	0.106	1.132		+0.003
32	0.120	0.110	1.091		-0.038
33	0.130	0.119	1.092		-0.037

There is insufficient data to look for trends with spectrum.

	Mean C/E	Error on Mean C/E $[\sum D^2/n(n-1)]^{1/2}$	Error on Individual Values $[\sum (D^2/n)]^{1/2}$
Mean of 6 (excl. 34)	1.129	0.015	0.034

Best value of C/E = 1.129 ± 0.015

(a) Rejected as being more than three standard deviations from the mean.

Table VII  
COMPARISON OF EXPERIMENTAL AND CALCULATED  
 $U^{238}/U^{235}$  FISSION RATIOS

Assembly Number	Calculated Ratio (C)	Experimental Ratio (E)	C/E	Analysis of Halves of Set		Analysis of Whole Set	
				Average C/E	Deviation (D)	Average C/E	Deviation (D)
25	0.0328	0.0292	1.123	1.153	-0.030	1.161	-0.039
38(a)	0.0343	0.0308	1.114		-0.039		-0.047
35	0.0367	0.0301	1.219		+0.066		+0.058
11	0.0404	0.0355	1.138		-0.015		-0.023
34	0.0407	0.0339	1.201		+0.048		+0.040
29	0.0409	0.0356	1.149		-0.004		-0.012
20	0.0444	0.0381	1.165		+0.012		+0.004
16	0.0460	0.0414	1.111		-0.042		-0.050
36	0.0475	0.0410	1.159		+0.006		-0.002
30	0.0493	0.0427	1.155	1.168	-0.013		-0.006
10	0.0495	0.0440	1.125		-0.043		-0.036
12	0.0506	0.0444	1.140		-0.028		-0.021
32	0.0509	0.0451	1.129		-0.039		-0.032
31	0.0517	0.0440	1.175		+0.007		+0.014
33	0.0565	0.0480	1.177		+0.009		+0.016
17	0.0571	0.0490	1.165		-0.003		+0.004
14	0.0681	0.0550	1.238		+0.070		+0.077
23	0.0818	0.0678	1.206		+0.038		+0.045
				Mean C/E		Error on Mean C/E $[\sum D^2/n(n-1)]^{1/2}$	
						Error on Individual Values $[\sum (D^2/n)]^{1/2}$	
First 9	1.153		0.013			0.036	
Last 9	1.168		0.012			0.035	
18	1.161		0.009			0.036	

There is no evidence of a trend with spectrum.

$$\text{Best value of } C/E = 1.161 \pm 0.009$$

(a) The Assembly 24 experimental value is rejected. The calculated ratios for 24 and 38 are identical.

Table VIII

AVERAGE VALUES OF THE RATIO OF CALCULATED AND  
MEASURED FISSION RATIOS CORRECTED FOR  
INELASTIC SCATTERING IN THE  
FISSION CHAMBER WALLS

Fission Ratio	Average Calculated Ratio Kirn Ratio C/E	Error on Individual Value of C/E	Inelastic Scattering Correction (Table I)	Corrected Calculated Ratio Experimental Ratio
$\frac{\text{Pu}^{239}}{\text{U}^{235}}$	$1.058 \pm 0.004$	1.5%	Zero	$1.06 \pm \frac{1}{2}\%$
$\frac{\text{U}^{233}}{\text{U}^{235}}$	$1.049 \pm 0.004$	1.2%	Zero	$1.05 \pm \frac{1}{2}\%$
$\frac{\text{U}^{234}}{\text{U}^{235}}$	$1.179 \pm 0.012$	4.9%	$4\% \pm 1\%$	$1.14 \pm 1\frac{1}{2}\%$
$\frac{\text{Pu}^{240}}{\text{U}^{235}}$	$1.102 \pm 0.020$	5.0%	$4\% \pm 1\%$	$1.06 \pm 2\%$
$\frac{\text{U}^{236}}{\text{U}^{235}}$	$1.129 \pm 0.015$	3.4%	$6\% \pm 1\frac{1}{2}\%$	$1.07 \pm 2\%$
$\frac{\text{U}^{238}}{\text{U}^{235}}$	$1.161 \pm 0.009$	3.6%	$8\% \pm 2\%$	$1.08 \pm 2\%$

Table IX

AVERAGE CORRECTED VALUES OF THE RATIO OF  
CALCULATED AND MEASURED FISSION RATIOS  
WHEN TAKEN RELATIVE TO  $\text{Pu}^{239}$

Fission Ratio	$\frac{\text{U}^{235}}{\text{Pu}^{239}}$	$\frac{\text{U}^{233}}{\text{Pu}^{239}}$	$\frac{\text{U}^{234}}{\text{Pu}^{239}}$	$\frac{\text{Pu}^{240}}{\text{Pu}^{239}}$	$\frac{\text{U}^{236}}{\text{Pu}^{239}}$	$\frac{\text{U}^{238}}{\text{Pu}^{239}}$
Corrected						
Calculated Ratio	0.94	0.99	1.08	1.00	1.01	1.02
Experimental Ratio	$\pm \frac{1}{2}\%$	$\pm \frac{1}{2}\%$	$\pm 1\frac{1}{2}\%$	$\pm 2\%$	$\pm 2\%$	$\pm 2\%$

## 5. DISCUSSION OF RESULTS

Examination of Tables II through IX leads to the following observations:

1. The most important observation is that the average calculated values of the relative fission rates of  $\text{Pu}^{239}$ ,  $\text{Pu}^{240}$ ,  $\text{U}^{233}$ ,  $\text{U}^{236}$ , and  $\text{U}^{238}$  are in good agreement with the experimental values, whereas those for  $\text{U}^{235}$  and  $\text{U}^{234}$  are, respectively, 6% low and 8% high. It should be noted that the



agreement with the threshold fissile materials  $\text{Pu}^{240}$ ,  $\text{U}^{236}$ , and  $\text{U}^{238}$  is evident only when the correction for inelastic scattering in the Kirn chamber walls is applied. In the case of  $\text{U}^{234}$  this correction reduces, but does not remove, the discrepancy between calculation and experiment.

2. The relative values of the calculated and measured  $\text{U}^{233}/\text{U}^{235}$ ,  $\text{U}^{234}/\text{U}^{235}$ , and  $\text{U}^{238}/\text{U}^{235}$  fission ratios are independent of the hardness of the spectrum. There is some evidence that the ratio of calculated to measured  $\text{Pu}^{239}/\text{U}^{235}$  fission ratio decreases as the spectra increase in hardness, but this may not be statistically significant. There are insufficient values of  $\text{Pu}^{240}/\text{U}^{235}$  and  $\text{U}^{236}/\text{U}^{235}$  fission ratios to look for any trends with varying spectra.

3. The spread of individual values of the ratio of calculated and measured fission ratios (see the third column of Table VIII) is about  $1\frac{1}{2}\%$  for  $\text{Pu}^{239}/\text{U}^{235}$  and  $\text{U}^{233}/\text{U}^{235}$ , but is about  $3\frac{1}{2}\%$  to  $5\%$  for the other ratios. The accuracies of all the experimental fission ratios, based solely upon counting statistics, is  $1\%$  to  $2\%$ .

The implications of these observations are, of course, entirely dependent upon the assumptions made about the accuracy of the fission chamber measurements, and it must be emphasized that the only direct corroborative evidence for their accuracy is the agreement between  $\text{U}^{238}/\text{U}^{235}$  ratios measured with both Kirn and Miles chambers (see Appendix II). Even here the same techniques are used in both cases so that there is a possibility of an unknown systematic error. However, the discussions in Section 2.4 did not show any good reasons for doubting the correctness of the measurements (after allowance for inelastic scattering in the chamber walls), and we will therefore examine the observations in terms of the assumption that the experimental data are valid. We will also try to show how any conclusions are dependent upon the correctness of the measurements.

We first consider observation 1:

If the experimental data are correct, then the calculated relative fission rates of  $\text{Pu}^{239}$ ,  $\text{Pu}^{240}$ ,  $\text{U}^{233}$ ,  $\text{U}^{236}$ , and  $\text{U}^{238}$  are correct. Hence, as  $\text{Pu}^{239}$  and  $\text{U}^{233}$  are fissile over the entire energy range, whereas the other isotopes have thresholds at three different energies, the simplest conclusion is that the average values of the assumed fission cross sections of these five isotopes are fairly accurate and also that the calculated spectra are not greatly in error. It then follows that the average values of the assumed fission cross sections of  $\text{U}^{235}$  and  $\text{U}^{234}$  are, respectively,  $6\%$  too low and  $8\%$  too high. It should be noted that it is possible that there are considerable errors in both the fission cross sections and the calculated spectra, and that these fortuitously cancel to give the correct reaction rates, but this does not seem very probable, as the spectra in the 18 assemblies are determined by the nuclear properties of a considerable number of isotopes.

However, it must be remembered that the threshold fissile materials only give information regarding the fractions of flux above and below the thresholds, and thus they do not provide enough information to define neutron spectra accurately. In particular, they cannot give information on the shape of the low-energy end of the spectrum.

Some further deductions regarding the accuracy of the calculated spectra are made below in the examination of observation 3. If some or all of the experimental data are in error, then radically different conclusions can be derived from 1. For example, we could accept the correctness of the  $\text{Pu}^{239}$ ,  $\text{Pu}^{240}$ ,  $\text{U}^{233}$ ,  $\text{U}^{236}$ , and  $\text{U}^{238}$  measurements and also accept the above inferences regarding the accuracy of their fission cross sections and the reactor spectra; then we could attribute the discrepancies with  $\text{U}^{235}$  and  $\text{U}^{234}$  to errors in the quantities of fissile material in their respective fission chambers. If this were true, then all the calculated and measured fission ratios would be in fair agreement, but the evidence of the  $\text{U}^{238}/\text{U}^{235}$  measurements with Kirn and Miles chambers does not support this hypothesis, and there is no real evidence of its correctness.

The agreement between calculated and measured rates for  $\text{Pu}^{239}$ ,  $\text{Pu}^{240}$ ,  $\text{U}^{233}$ ,  $\text{U}^{236}$ , and  $\text{U}^{238}$  is circumstantial evidence of the correctness of those experimental measurements, the fission cross sections, and the calculated spectra, but it is interesting to speculate on the conclusions which would be reached if it were found that the  $\text{Pu}^{239}$  and  $\text{U}^{233}$  fission chambers were incorrectly calibrated. Under these circumstances, which are by no means incredible, it is possible that the calculated and measured fission rates of  $\text{U}^{233}$ ,  $\text{Pu}^{239}$ , and  $\text{U}^{235}$  could be in agreement. If the data on the other fissile materials remained unchanged, then one would possibly deduce that the calculated fission rates of the threshold fissile materials were in error due to incorrectly calculated spectra.

Clearly, there are considerable uncertainties if it is accepted that certain of the fission chambers may not contain the anticipated quantity of fissile material, and interpretation of fission ratio measurements would be greatly simplified if the intercalibration of the fission chambers could be established unambiguously.

Next we examine observation 2:

This gives some information about the accuracies of the assumed variations of the fission cross sections with neutron energy since, if these are in error, the relative values of the calculated and measured fission ratios will alter as the spectrum is progressively hardened or softened. Thus, the indications are that there are no gross errors in the assumed variations of the  $\text{U}^{233}$ ,  $\text{U}^{234}$ ,  $\text{U}^{235}$ , and  $\text{U}^{238}$  cross sections, and that there is some evidence that the assumed variation for  $\text{Pu}^{239}$  is slightly incorrect. These inferences are not dependent upon the correctness of the fission chamber measurements, as the only likely source of error in these is in

their relative sensitivities and this would not affect the variations of fission ratios with changing spectrum. It should be noted that, since the differences in spectra in the assemblies examined are not very great, and since only average cross sections are concerned, this type of examination will only reveal errors if these are large.

Finally we consider observation 3:

The statistical accuracies of all the measured fission ratios are about 1% to 2%. The same accuracy would be obtained in the ratio of calculated and measured ratios if there were no errors in the calculated data. In fact, we see that spreads of  $3\frac{1}{2}\%$  to 5% are obtained with the ratios which include threshold fissile materials, and this indicates that there are uncertainties of this order in the calculated fractions of the fluxes lying above the threshold energies. The uncertainties are attributed to spectra, rather than cross sections, as the latter are fairly flat above the threshold energies.

The  $U^{233}/U^{235}$  and  $Pu^{239}/U^{235}$  data only exhibit the experimental uncertainty of about  $1\frac{1}{2}\%$  because they are rather insensitive to errors in spectrum.

## 6. SUMMARY OF CONCLUSIONS AND POSSIBLE FUTURE WORK

This study shows that, when the measured data for threshold fissile materials are corrected for the effects of inelastic scattering in the fission chamber walls, calculated and measured fission rates for a number of isotopes are in fair agreement.

The relative fission rates of  $Pu^{239}$  and  $U^{233}$  can be calculated to about  $\pm 1\frac{1}{2}\%$  and those of  $Pu^{240}$ ,  $U^{236}$ , and  $U^{238}$  relative to either  $Pu^{239}$  or  $U^{233}$  can be calculated to about  $\pm 3\frac{1}{2}\%$  to  $\pm 5\%$ . The larger uncertainty with the threshold isotopes is probably due to uncertainties of the order of  $\pm 5\%$  in the calculated fraction of flux above the threshold energies.

The calculated fission rates of  $U^{235}$  and  $U^{234}$ , however, are about 6% low and 8% high, respectively, relative to those of the other five isotopes.

If the measurements are all valid, it appears that the calculated spectra are moderately correct and that the assumed fission cross sections of  $Pu^{239}$ ,  $Pu^{240}$ ,  $U^{233}$ ,  $U^{236}$ , and  $U^{238}$  are fairly accurate. The assumed fission cross sections of  $U^{235}$  and  $U^{234}$  would then be in error, being 6% low and 8% high, respectively. However, the correctness of these conclusions depends strongly upon the accuracy of the fission chamber measurements, and, although the design and manufacture of the fission chambers are such that the measurements should be good, there is little independent evidence for their accuracy.

The discussion in Section 5 shows that the discrepancies with  $U^{235}$  and  $U^{234}$  could be due to erroneous intercalibration of the  $U^{235}$  and  $U^{234}$  chambers relative to the others. Also, this discussion indicates that quite different conclusions could be reached if the  $U^{233}$  and  $Pu^{239}$  chamber measurements were in error.

Clearly, it is important to be certain of the intercalibration of the fission chambers, and it would be particularly useful if an independent calibration could be made. A possible technique with the chambers which contain thermally fissile material might be the construction of Kirn-type chambers whose overall efficiencies could be checked by counting in a thermal flux.

Additional experiments with thick- and thin-walled chambers with a variety of fissile materials in a number of assemblies would also be valuable in order to establish the magnitude of the effects of inelastic scattering more accurately.

#### ACKNOWLEDGEMENTS

The author was greatly helped in this work by having access to original experimental data and would like to thank all the ZPR-III staff for their cooperation in this matter. In addition, he would like to thank J. M. Chaumont for certain DSN calculations, and G. J. Fischer for the fission cross sections of  $U^{234}$  and  $U^{236}$ .

Particular thanks are due to R. R. Smith, F. S. Kirn, and C. C. Miles for many stimulating conversations and a wealth of information on the manufacture and testing of fission chambers.

Finally, he would like to thank F. W. Thalgot for his support and encouragement of this study.

## REFERENCES

1. S. Yiftah, D. Okrent, and P. A. Moldauer, Fast Reactor Cross Sections, Pergamon Press, New York (1960).
2. J. K. Long et al., Experimental Results on Large Dilute Fast Critical Systems with Metallic and Ceramic Fuels, Paper presented at the IAEA Sponsored Seminar on the Physics of Fast and Intermediate Reactors, Vienna (1961), Paper No. SM-18/48.
3. W. G. Davey and R. N. Curran, An Experimental Investigation of Some Sources of Error in the Measurement of Absolute Fission Ratios in Fast Reactors, ANL-6468 (Nov 1961).
4. F. S. Kirn, An Absolute Fission Counter, American Nuclear Society, 2nd Winter Meeting (1957).
5. J. K. Long et al., Fast Neutron Power Reactor Studies with ZPR-III, Proceedings of the Second UN International Conference on the Peaceful Uses of Atomic Energy, Geneva, Switzerland, 12, 119 (1958).
6. W. G. Davey, k Calculations for 22 ZPR-III Fast Reactor Assemblies Using ANL Cross-section Set 635, ANL-6570 (May 1962).
7. T. A. Doyle and A. L. Hess, Critical Studies of Core B for the Enrico Fermi Atomic Power Plant (ZPR-III Assembly 35), To be published.
8. J. C. Bates, W. G. Davey, P. I. Amundson, J. M. Gasidlo, J. K. Long, and W. P. Keeney, Two Spherical Fast Critical Experiments (ZPR-III Assemblies 38 and 39), ANL-6582 (July 1962).
9. D. Meneghetti and M. F. Loomis, Calculation of Heterogeneity Effects in ZPR-III Fast Assemblies Using the DSN Program, ANL-6218 (Nov 1960).
10. A. L. Hess, W. Gemmell, J. K. Long, and R. L. McVean, Critical Studies of a 450-liter Uranium Oxide Fast Reactor Core (ZPR-III Assembly 29), ANL-6336 (May 1961).
11. P. I. Amundson, A. L. Hess, W. P. Keeney, J. K. Long, and R. L. McVean, Critical Studies of a Dilute Oxide Fast Reactor Core (ZPR-III Assembly 30), ANL-6337 (May 1961).
12. J. M. Gasidlo, J. K. Long, and R. L. McVean, Critical Studies of a Dilute Fast Reactor Core (ZPR-III Assembly 31), ANL-6338 (Oct 1961).
13. R. J. Huber, J. K. Long, R. L. McVean, and J. M. Gasidlo, Critical Studies of a Dilute Carbide Fast Reactor Core (ZPR-III Assembly 34), ANL-6401 (May 1961).
14. J. M. Gasidlo, J. K. Long, and R. L. McVean, Critical Studies of a Fast Reactor Core Containing Depleted Uranium and Sodium as Diluents (ZPR-III Assembly 36), ANL-6401 (Jan 1962).



## APPENDIX I

Perturbation of Fission Ratios by Fission Chambers  
and Steel Shells

A. Measurements in ZPR-III Assemblies 35(7) and 38(8)

These measurements investigated both the effect of chamber wall thickness and the effect of adding an extra 3-in. length of cable behind the chamber. Kirn and Miles HWC and TWC chambers were used (see Fig. 1 for details of these chambers). Results are given in the following table:

Experiment Number	Assembly	Chamber	Extra 3 in. of Cable	Fission Ratio
1	35	TWC	No	$U^{238}/U^{235}$ 0.0314 $\pm$ 0.0002
2	35	TWC	Yes	$U^{238}/U^{235}$ 0.0309 $\pm$ 0.0002
3	35	HWC	No	$U^{238}/U^{235}$ 0.0290 $\pm$ 0.0003
4	38	Kirn	No	$U^{238}/U^{235}$ 0.0308 $\pm$ 0.0003
5	38	Kirn	Yes	$U^{238}/U^{235}$ 0.0308 $\pm$ 0.0003
6	38	Kirn	No	$U^{234}/U^{235}$ 0.268 $\pm$ 0.003
7	38	Kirn	Yes	$U^{234}/U^{235}$ 0.264 $\pm$ 0.003
8	38	HWC	No	$U^{238}/U^{235}$ 0.0312 $\pm$ 0.0003
9	38	TWC	No	$U^{238}/U^{235}$ 0.0335 $\pm$ 0.0003

Comparison of experiments 1 with 2, 4 with 5, and 6 with 7 gives the effect of adding extra cable (and hence gives the effect of the cable already present). It can be seen that there is no evidence that the cable affects either the  $U^{238}/U^{235}$  or the  $U^{234}/U^{235}$  fission ratios.

Comparison of 1 with 3 and of 8 with 9 gives the effect of the chamber walls on the  $U^{238}/U^{235}$  ratio. It can be seen that there is a  $8.3\% \pm 1.0\%$  effect in Assembly 35 and a  $7.4\% \pm 1.5\%$  effect in Assembly 38. We conclude that the chamber walls alter the  $U^{238}/U^{235}$  fission ratio by 8% in both assemblies. This supports the calculations in Section C of the Appendix, which indicate that the effect of an iron shell on fission ratios is fairly independent of the composition of the reactor core.

B. Measurements in AFSR<sup>(3)</sup>

Measurements were made in the grazing hole of AFSR, this hole being just outside the reactor core. The experiments consisted of measuring the effect of placing thick-walled steel cylinders around  $U^{238}/U^{235}$ ,  $U^{234}/U^{235}$ , and  $Pu^{239}/U^{235}$  fission chamber pairs.

The chambers were cylindrical with an OD of about 1 cm and an active length of about 3 cm, and the steel cylinders were of 1.23-cm ID and 2.5-cm OD (i.e., 0.635-cm wall thickness). The results were as follows:

<u>Fission Ratio</u>	<u>Percentage Change on Adding the Steel Cylinder</u>
$U^{238}/U^{235}$	$-4.2 \pm 0.5$
$U^{234}/U^{235}$	$-2.6 \pm 0.5$
$Pu^{239}/U^{235}$	$0.0 \pm 0.2$

The negative sign indicates that a fission ratio decreases on addition of the steel cylinder.

The magnitude of the change in the  $U^{238}/U^{235}$  ratio in these experiments is about half that found in the ZPR-III experiments (see Section A of this Appendix), but this is quite reasonable, as the conditions of the two experiments are grossly different. The sign of the change is of course the same.

#### C. DSN Calculations for Assemblies 14, 25, and 32

All these calculations were carried out with the Yiftah, Okrent, and Moldauer ANL Set 135, the DSN neutron transport code being used.

In each case, two calculations were performed: (a) with a 2.058-cm-radius void at the core center, and (b) with a 1.708-cm void and an iron shell of 1.708-cm inner radius and 2.058-cm outer radius (i.e., 0.35-cm wall thickness) at the core center. From these two calculations the central spectra and central fission cross sections and ratios were obtained, thus obtaining the change in fission ratios due to addition of the iron shell.

The three Assemblies 14, 25, and 32 were chosen for these calculations so that there is a considerable variation of core composition and of hardness of spectrum. All three assemblies were examined with the same mesh at the core center and the S4 approximation; in addition, the Assembly 32 calculation was repeated with a finer mesh and the S8 approximation.

The core composition and results of the calculations are given in Tables I and II.

Table I

#### REACTOR DIMENSIONS AND COMPOSITIONS

<u>Assembly</u>	<u>Outer Core Radius (cm)</u>	<u>Outer Blanket Radius (cm)</u>	<u>Core Composition (v/o)</u>			
			<u>U<sup>235</sup></u>	<u>U<sup>238</sup></u>	<u>Fe</u>	<u>C</u>
14	25.9	55.9	0.0938	0.0070	0.0922	0.6392
25	48.0	88.0	0.0717	0.7417	0.0928	-
32	32.0	62.0	0.0926	0.0066	0.8100	-



Table II

CALCULATED PERCENTAGE CHANGES IN CENTRAL FISSION  
CROSS SECTIONS AND FISSION RATIOS DUE TO THE  
ADDITION OF A 0.35-cm WALL IRON SHELL

Parameter	Assembly 14 (S4)	Assembly 25 (S4)	Assembly 32 (S4)	Assembly 32 (S8)
$\sigma_f \text{U}^{235}$	+0.16	+0.20	+0.14	+0.14
$\sigma_f \text{U}^{233}$	+0.19	+0.19	+0.15	+0.15
$\sigma_f \text{Pu}^{239}$	+0.01	+0.02	-0.01	-0.01
$\sigma_f \text{Pu}^{240}$	-0.83	-0.92	-0.87	-0.87
$\sigma_f \text{U}^{234}$	-0.78	-0.85	-0.80	-0.80
$\sigma_f \text{U}^{236}$	-1.56	-1.62	-1.60	-1.60
$\sigma_f \text{U}^{238}$	-2.19	-2.17	-2.25	-2.25
$\frac{\sigma_f \text{U}^{233}}{\sigma_f \text{U}^{235}}$	+0.03	-0.01	+0.02	+0.02
$\frac{\sigma_f \text{Pu}^{239}}{\sigma_f \text{U}^{235}}$	-0.15	-0.18	-0.15	-0.15
$\frac{\sigma_f \text{Pu}^{240}}{\sigma_f \text{U}^{235}}$	-0.99	-1.12	-1.01	-1.01
$\frac{\sigma_f \text{U}^{234}}{\sigma_f \text{U}^{235}}$	-0.94	-1.05	-0.94	-0.94
$\frac{\sigma_f \text{U}^{236}}{\sigma_f \text{U}^{235}}$	-1.72	-1.82	-1.73	-1.73
$\frac{\sigma_f \text{U}^{238}}{\sigma_f \text{U}^{235}}$	-2.34	-2.37	-2.38	-2.38

NOTE: A positive sign indicates an increase on addition of the iron shell.

The data given in Table II show that the calculated changes in average fission cross sections and fission ratios are comparatively insensitive to the reactor core considered or to the order of the Sn calculation. It therefore appears justifiable to assume that the correction to a measured fission ratio for the effects of the chamber wall is the same for all the ZPR-III Assemblies.

The differences between the magnitude of the calculated changes and those measured in ZPR-III (see Section A, Appendix I) is to be expected, as the geometries and wall thickness differ considerably.



## APPENDIX II

Experimental Evidence for the Validity of  
Fission Chamber Measurements

A. Intercomparison of Kirn  $U^{235}$  Chambers 4 and 5Assumed U mass in Chamber 4    802  $\mu\text{gm}$ Assumed U mass in Chamber 5    804  $\mu\text{gm}$ 

The chambers were manufactured with use of the same uranium solution. The uranium was 93.41 w/o  $U^{235}$ , 5.52 w/o  $U^{238}$ , and 1.07 w/o  $U^{234}$ .

Expected Ratio of Count Rates,  $\frac{4}{5} = \frac{802}{804} = 0.998$ .

Measured Ratio of Count Rates as follows:

ZPR-III Assembly 30	$1.001 \pm 0.005$
ZPR-III Assembly 31	$0.981 \pm 0.005$
ZPR-III Assembly 31	$1.017 \pm 0.005$
ZPR-III Assembly 34	$0.989 \pm 0.005$
Mean Measured Ratio =	$0.997 \pm 0.008$

Thus there is excellent agreement between the measured relative count rates and those predicted on the basis of the mass of uranium deposited.

B.  $U^{238}/U^{235}$  Fission Ratios Measured with Kirn and Miles Chambers

The Kirn and Miles chambers were manufactured by different people using different apparatus and uranium solutions of different isotopic composition. Some years also lapsed between the manufacture of the two types. The  $U^{238}/U^{235}$  ratio measured with the Kirn chambers should, however, be closely similar to that measured with the Miles Heavy Wall Chambers (HWC). Figure 1 shows the similarity of Kirn and HWC.

	$U^{238}/U^{235}$ Fission Ratio	
	Kirn	Miles HWC
Assembly 35	$0.0296 \pm 0.0003$	$0.0290 \pm 0.0003$
Assembly 38	$0.0308 \pm 0.0003$	$0.0312 \pm 0.0003$

The errors are one standard deviation, and hence it can be seen that the Kirn and Miles HWC give identical results. It therefore seems probable that the measured ratios are correct.

## APPENDIX I

Table 1. Summary of the results of the experiments conducted during the summer of 1964.

## Experiment 1

Experiment 1. Summary of results.

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Experiment 1. Summary of results.

## APPENDIX III

Group Fission Cross Sections for  $U^{234}$  and  $U^{236}$  (ANL Set 179)

<u>Group</u>	<u><math>\sigma_f U^{234}</math> (b)</u>	<u><math>\sigma_f U^{236}</math> (b)</u>
1	1.50	0.98
2	1.47	0.91
3	1.41	0.73
4	1.20	0.41
5	0.793	0.035
6	0.290	
7	0.059	
8	0.029	
9		
10		
11		
12		
13		
14		
15		
16		



## APPENDIX IV

## Central Spectra and Fission Ratios

Assembly																		
Group	10	11	12	14	16	17	20	23	24	25	29	30	31	32	33	34	35	36
Central Spectra																		
1	0.024	0.020	0.026	0.036	0.023	0.030	0.020	0.036	0.017	0.016	0.019	0.021	0.022	0.019	0.021	0.018	0.015	0.023
2	0.041	0.034	0.048	0.074	0.041	0.057	0.038	0.072	0.029	0.028	0.039	0.044	0.044	0.041	0.046	0.038	0.033	0.040
3	0.065	0.053	0.073	0.112	0.064	0.086	0.065	0.112	0.046	0.044	0.066	0.076	0.077	0.080	0.088	0.067	0.065	0.066
4	0.121	0.109	0.112	0.118	0.111	0.114	0.111	0.144	0.099	0.097	0.100	0.114	0.123	0.125	0.128	0.107	0.096	0.115
5	0.204	0.199	0.156	0.118	0.173	0.140	0.169	0.162	0.193	0.191	0.141	0.160	0.174	0.195	0.186	0.148	0.143	0.193
6	0.207	0.212	0.155	0.108	0.177	0.134	0.185	0.156	0.214	0.214	0.146	0.162	0.179	0.187	0.186	0.155	0.167	0.206
7	0.147	0.157	0.130	0.096	0.143	0.117	0.151	0.120	0.163	0.164	0.140	0.141	0.139	0.137	0.135	0.132	0.146	0.151
8	0.098	0.108	0.102	0.080	0.107	0.094	0.107	0.080	0.116	0.118	0.108	0.102	0.096	0.092	0.092	0.102	0.113	0.104
9	0.046	0.048	0.074	0.072	0.067	0.076	0.071	0.062	0.052	0.054	0.087	0.076	0.068	0.062	0.061	0.082	0.084	0.050
10	0.034	0.043	0.057	0.058	0.052	0.059	0.053	0.038	0.050	0.052	0.072	0.057	0.049	0.037	0.034	0.069	0.059	0.038
11	0.007	0.008	0.030	0.041	0.021	0.037	0.016	0.011	0.010	0.011	0.033	0.022	0.015	0.011	0.011	0.033	0.027	0.009
12	0.005	0.007	0.020	0.032	0.013	0.026	0.011	0.006	0.009	0.009	0.028	0.017	0.011	0.009	0.008	0.028	0.026	0.006
13	0.001	0.002	0.010	0.022	0.005	0.015	0.002	0.001	0.002	0.002	0.011	0.005	0.002	0.003	0.002	0.011	0.011	0.002
14			0.004	0.014	0.002	0.008	0.001				0.005	0.002	0.001	0.001	0.001	0.005	0.006	
15			0.002	0.011	0.001	0.005					0.004	0.001		0.001	0.001	0.004	0.006	
16			0.001	0.008		0.002					0.001					0.001	0.003	
Average Central Cross Sections (b)																		
$\sigma_f^{235}\text{U}$	1.399	1.420	1.537	1.701	1.480	1.596	1.462	1.411	1.438	1.443	1.591	1.497	1.450	1.431	1.424	1.582	1.595	1.414
$\sigma_f^{233}\text{U}$	2.219	2.253	2.379	2.553	2.317	2.444	2.296	2.200	2.281	2.288	2.455	2.333	2.274	2.255	2.242	2.445	2.465	2.240
$\sigma_f^{239}\text{Pu}$	1.765	1.763	1.815	1.909	1.788	1.846	1.779	1.796	1.763	1.763	1.825	1.797	1.784	1.780	1.782	1.824	1.827	1.767
$\sigma_f^{240}\text{Pu}$	0.5759	0.5202	0.5435	0.6324	0.5295	0.5711	0.5203	0.7089	0.4784	0.4678	0.4776	0.5417	0.5711	0.5866	0.6076	0.4924	0.4602	0.5529
$\sigma_f^{234}\text{U}$	0.5664	0.5172	0.5262	0.5952	0.5183	0.5464	0.5104	0.6737	0.4799	0.4702	0.4644	0.5253	0.5547	0.5712	0.5889	0.4788	0.4516	0.5488
$\sigma_f^{236}\text{U}$	0.1650	0.1409	0.1738	0.2369	0.1581	0.1957	0.1531	0.2473	0.1240	0.1197	0.1482	0.1684	0.1743	0.1724	0.1857	0.1502	0.1365	0.1588
$\sigma_f^{238}\text{U}$	0.0692	0.0573	0.0778	0.1159	0.0680	0.0911	0.0649	0.1154	0.0494	0.0473	0.0650	0.0738	0.0750	0.0728	0.0804	0.0644	0.0586	0.0671
Central Fission Ratios																		
$\sigma_f^3/\sigma_f^5$	1.587	1.587	1.548	1.501	1.566	1.532	1.571	1.559	1.586	1.586	1.543	1.559	1.569	1.576	1.574	1.545	1.545	1.584
$\sigma_f^9/\sigma_f^5$	1.262	1.242	1.181	1.122	1.208	1.156	1.217	1.273	1.226	1.222	1.147	1.201	1.231	1.244	1.251	1.153	1.145	1.250
$\sigma_f^0/\sigma_f^5$	0.412	0.366	0.354	0.372	0.358	0.358	0.356	0.502	0.333	0.324	0.300	0.362	0.394	0.410	0.427	0.311	0.288	0.391
$\sigma_f^4/\sigma_f^5$	0.405	0.364	0.342	0.350	0.350	0.342	0.349	0.477	0.334	0.326	0.292	0.351	0.383	0.399	0.414	0.303	0.283	0.385
$\sigma_f^6/\sigma_f^5$	0.118	0.0992	0.113	0.139	0.107	0.123	0.105	0.175	0.0862	0.0830	0.0931	0.112	0.120	0.120	0.130	0.0949	0.0856	0.112
$\sigma_f^8/\sigma_f^5$	0.0495	0.0404	0.0506	0.0681	0.0460	0.0571	0.0444	0.0818	0.0343	0.0328	0.0409	0.0493	0.0517	0.0509	0.0565	0.0407	0.0367	0.0475





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